JC19 Rec'd PCT/PTO 01 JUN 2001

FORM PTO-1390 (Modified) (REV 11-98) U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371

ATTORNEY'S DOCKET NUMBER
JMYT-245US

U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR 1.5)
To Be Assigned 09/857116

INTERNATIONAL APPLICATION NO.	INTERNATIONAL FILING DATE	PRIORITY DATE CLAIMED
PCT/GB99/03958	**29 November 1999 (29.11.99)**	**01 December 1998 (01.12.98)**

TITLE OF INVENTION
REACTOR AND PROCESS FOR REMOVAL OF CARBON MONOXIDE FROM HYDROGEN

APPLICANT(S) FOR DO/EO/US
GRAY, Peter Geoffrey
PIGNON, John Frederick

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☒ This is an express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).
4. ☒ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
5. ☒ A copy of the International Application as filed (35 U.S.C. 371 (c) (2))
   a. ☒ is transmitted herewith (required only if not transmitted by the International Bureau).
   b. ☐ has been transmitted by the International Bureau.
   c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☐ A translation of the International Application into English (35 U.S.C. 371(c)(2)).
7. ☒ A copy of the International Search Report (PCT/ISA/210).
8. ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371 (c)(3))
   a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
   b. ☐ have been transmitted by the International Bureau.
   c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
   d. ☒ have not been made and will not be made.
9. ☐ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
10. ☒ An oath or declaration of the inventor(s) (35 U.S.C. 371 (c)(4)). (UNEXECUTED)
11. ☒ A copy of the International Preliminary Examination Report (PCT/IPEA/409).
12. ☐ A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371 (c)(5)).

Items 13 to 20 below concern document(s) or information included:

13. ☐ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
14. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
15. ☒ A **FIRST** preliminary amendment.
16. ☐ A **SECOND** or **SUBSEQUENT** preliminary amendment.
17. ☐ A substitute specification.
18. ☐ A change of power of attorney and/or address letter.
19. ☒ Certificate of Mailing by Express Mail
20. ☐ Other items or information:

PCTUS1/REV03

U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR 1.5)	INTERNATIONAL APPLICATION NO.	ATTORNEY'S DOCKET NUMBER
09/857116 To Be Assigned	PCT/GB99/03958	JMYT-245US

21. The following fees are submitted:.

	CALCULATIONS	PTO USE ONLY
BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)) :		
☐ Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2) paid to USPTO and International Search Report not prepared by the EPO or JPO $970.00		
☒ International preliminary examination fee (37 CFR 1.482) not paid to USPTO but Internation Search Report prepared by the EPO or JPO $840.00		
☐ International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO $690.00		
☐ International preliminary examination fee paid to USPTO (37 CFR 1.482) but all claims did not satisfy provisions of PCT Article 33(1)-(4) $670.00		
☐ International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(1)-(4) $96.00		
ENTER APPROPRIATE BASIC FEE AMOUNT =	$860.00	
Surcharge of **$130.00** for furnishing the oath or declaration later than ☐ 20 ☐ 30 months from the earliest claimed priority date (37 CFR 1.492 (e)).	$0.00	

CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE		
Total claims	14 - 20 =	0	x $18.00	$0.00	
Independent claims	2 - 3 =	0	x $80.00	$0.00	
Multiple Dependent Claims **(check if applicable).**			☐	$0.00	
	TOTAL OF ABOVE CALCULATIONS		=	$860.00	
Reduction of 1/2 for filing by small entity, if applicable. Verified Small Entity Statement must also be filed (Note 37 CFR 1.9, 1.27, 1.28) **(check if applicable).**			☐	$0.00	
	SUBTOTAL		=	$860.00	
Processing fee of **$130.00** for furnishing the English translation later than ☐ 20 ☐ 30 months from the earliest claimed priority date (37 CFR 1.492 (f)).			+	$0.00	
	TOTAL NATIONAL FEE		=	$860.00	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31) **(check if applicable).**			☐	$0.00	
	TOTAL FEES ENCLOSED		=	$860.00	
				Amount to be: refunded	$
				charged	$

☒ A check in the amount of **$860.00** to cover the above fees is enclosed.

☐ Please charge my Deposit Account No. in the amount of to cover the above fees. A duplicate copy of this sheet is enclosed.

☒ The Commissioner is hereby authorized to charge any fees which may be required, or credit any overpayment to Deposit Account No. **18-0350** A duplicate copy of this sheet is enclosed.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:

Christopher R. Lewis
RATNER & PRESTIA
Suite 301
One Westlakes, Berwyn
P.O. Box 980
Valley Forge, PA 19482-0980

Telephone: (610) 407-0700
Facsimile: (610) 407-0701

SIGNATURE

Christopher R. Lewis
NAME

36,201
REGISTRATION NUMBER

June 1, 2001
DATE

09/857116

JC18 Rec'd PCT/PTO 01 JUN 2001

CERTIFICATE OF MAILING BY "EXPRESS MAIL" (37 CFR 1.10) Applicant(s): **Peter Geoffrey Gray et al.**		Docket No. **JMYT-245US**	
Serial No. **To Be Assigned**	Filing Date **Herewith**	Examiner	Group Art Unit

Invention: **REACTOR AND PROCESS FOR REMOVAL OF CARBON MONOXIDE FROM HYDROGEN**

I hereby certify that the following correspondence:

U.S. National Phase Application, w/Form PTO-1390 and all of the documents listed therein,

(Identify type of correspondence)

is being deposited with the United States Postal Service "Express Mail Post Office to Addressee" service under 37 CFR 1.10 in an envelope addressed to: The Assistant Commissioner for Patents, Washington, D.C. 20231

June 1, 2001
(Date)

Kathleen Libby
(Typed or Printed Name of Person Mailing Correspondence)

Kathleen Libby
(Signature of Person Mailing Correspondence)

EL732291825US
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09/857116
JC18 Rec'd PCT/PTO 01 JUN 2001

JMYT-245US

PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant:	Peter Geoffrey Gray et al.	: Art Unit:
Application No.:	To Be Assigned	: Examiner:
Filed:	Herewith	:
FOR: REACTOR AND PROCESS FOR REMOVAL OF CARBON MONOXIDE FROM HYDROGEN		:

PRELIMINARY AMENDMENT

Assistant Commissioner for Patents
Washington, DC 20231

SIR:

Prior to examination, please amend the above-identified application as follows.

IN THE SPECIFICATION:

Please replace the paragraph beginning at page 3, line 9, with the following:

An essentially cylindrical metal vessel, made by pressing two halves and seam-welding, has a main gas inlet, 2, for reformate gas, and an outlet, 3, for cleaned gas. The vessel has a number of annular reaction zones, 4, each consisting of an annular wound corrugated catalyst support. The vessel further has an internal water jacket, 5, between a spacer and the shell and this is supplied with cooling water by means of a water circuit comprising a circulating pump, 6, and a heat exchanger, 7. The heat

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exchanger is desirably used to recover heat in an integrated system e.g. in an integrated combined heat and power system. The vessel further is provided with four air inlets, 8, each providing pre-determined (for a steady-state operation) levels of air (that is reaction oxygen; air is generally suitable). Although not preferred, controlled amounts of oxygen, according to one or more feedback or feed forward controls detecting gas flow levels and CO levels may be used. Downstream of each air inlet is a gas mixing zone, 9a, 9b, 9c and 9d. Zone 9a is suitably a static mixer, but zones 9b, 9c and 9d are preferably alternating annular mixing vanes, 10, and discs, 11, having gas flow holes, as illustrated in Figs 2 and 3. It should be noted that for simplicity only four vanes and four holes are shown, but more, e.g. six or eight, may be used. Such vanes and discs may be manufactured by pressing to form a sleeve which may be fitted and brazed or welded onto the jacket 5 with the intention that each gas mixing zone also serves to remove heat from the reaction gases leaving each reaction zone. Although the schematic drawing of the reactor shows each reaction zone and gas mixing zone of the same size, these may be designed to differ according to the amount of heat to be removed. Additionally, each reaction zone may incorporate different and/or different loadings, of catalyst. Each metal catalyst support monolith suitably has 300-400 cells per sq.in. and is coated with a washcoat comprising ~36wt% alumina solids of D50 of approx $5\mu m$ and D90 of approx $16\mu m$, to deposit approx $3g/m^3$. After drying and firing, the coated monolith was impregnated with a platinum salt solution to result in 5% by wt of Pt deposited on the alumina. The resulting coated monolith was again dried and fired before being assembled into the reactor.

Please replace the paragraph beginning at page 5, line 1, with the following:

The detailed operating conditions and other experimental detail for one typical run of the above reactor now follow. It was remarkable that the pressure drop across the whole of the test reactor was about the

measurement limit of the gauges being used, and was of the order of 1.5-3mbar (150-300Pa), at the two throughputs tested. This indicates that the reactor is very efficient, compact and requires very little energy to operate.

IN THE CLAIMS:

Please replace claims 1, 3, 4, 6-8, 10, 11, and 14 with the following amended claims:

1. A catalytic selective oxidation reactor, comprising a cylindrical reactor vessel with counter current cooling means and at least one stage, each stage being provided with an inlet for a first feedstock, and inlet for a second feedstock, gas mixing means and a catalytic reaction zone, wherein the reaction zone comprises a selective oxidation catalyst deposited upon a metal support.

3. A reactor according to claim 1, wherein the metal support is a metal monolith.

4. A reactor according to claim 1, wherein the cooling means comprises a central jacket mounted within the reactor.

6. A reactor according to claim 4, wherein the central jacket is connected via a coolant circuit to a circulating pump.

7. A reactor according to claim 1, wherein the catalytic reaction zone(s) is/are annular and surround a central cooling means.

8. A reactor according to claim 1, having from 2 to 10 stages.

10. A reactor according to claim 1, wherein the gas mixing means comprise annular mixing vanes or discs.

11. A reactor according to claim 1, connected to the output from a reformer such that CO is removed from the output gas to a level where the output from the reactor may be fed to a fuel cell.

14. A process according to claim 12, providing hydrogen to a fuel cell.

Respectfully submitted,

Christopher R. Lewis, Reg. No. 36,201
Attorney for Applicants

CRL/lrb

Dated: June 1, 2001

Suite 301
One Westlakes, Berwyn
P.O. Box 980
Valley Forge, PA 19482-0980
(610) 407-0700

The Assistant Commissioner for Patents is hereby authorized to charge payment to Deposit Account No. **18-0350** of any fees associated with this communication.

EXPRESS MAIL Mailing Label No.: EL732291825US
Date of Deposit: June 1, 2001

I hereby certify that this paper and fee are being deposited, under 37 C.F.R. § 1.10 and with sufficient postage, using the "Express Mail Post Office to Addressee" service of the United States Postal Service on the date indicated above and that the deposit is addressed to the Assistant Commissioner for Patents, Washington, D.C. 20231.

Kathleen Libby

VERSION WITH MARKINGS TO SHOW CHANGES MADE

IN THE SPECIFICATION:

Specification at page 3, line 9:

An essentially cylindrical metal vessel, made by pressing two halves and seam-welding, has a main gas inlet, 2, for reformate gas, and an outlet, 3, for cleaned gas. The vessel has a number of annular reaction zones, 4, each consisting of an annular wound corrugated catalyst support. The vessel further has an internal water jacket, 5, between a spacer and the shell and this is supplied with cooling water by means of a water circuit comprising a circulating pump, 6, and a heat exchanger, 7. The heat exchanger is desirably used to recover heat in an integrated system e.g. in an integrated combined heat and power system. The vessel further is provided with four air inlets, 8, each providing pre-determined (for a steady-state operation) levels of air (that is reaction oxygen; air is generally suitable). Although not preferred, controlled amounts of oxygen, according to one or more feedback or feed forward controls detecting gas flow levels and CO levels may be used. Downstream of each air inlet is a gas mixing zone, 9a, 9b, 9c and 9d. Zone 9a is suitably a static mixer, but zones 9b, 9c and 9d are preferably alternating annular mixing vanes, 10, and discs, 11, having gas flow holes, as illustrated in Figs 2 and 3. It should be noted that for simplicity only four vanes and four holes are shown, but more, e.g. six or eight, may be used. Such vanes and discs may be manufactured by pressing to form a sleeve which may be fitted and brazed or welded onto the jacket 5 with the intention that each gas mixing zone also serves to remove ~~hear~~ heat from the reaction gases leaving each reaction zone. Although the schematic drawing of the reactor shows each reaction zone and gas mixing zone of the same size, these may be designed to differ according to the amount of heat to be removed. Additionally, each reaction zone may incorporate different

and/or different loadings, of catalyst. Each metal catalyst support monolith suitably has 300-400 cells per sq.in. and is coated with a washcoat comprising ~36wt% alumina solids of D50 of approx 5μm and D90 of approx 16μm, to deposit approx $3g/m^3$. After drying and firing, the coated monolith was impregnated with a platinum salt solution to result in 5% by wt of Pt deposited on the alumina. The resulting coated monolith was again dried and fired before being assembled into the reactor.

Specification at page 5, line 1:

The detailed operating conditions and other experimental detail for one typical run of the above reactor now follow. It was remarkable that the pressure drop across the whole of the ~~est~~ test reactor was about the measurement limit of the gauges being used, and was of the order of 1.5-3mbar (150-300Pa), at the two throughputs tested. This indicates that the reactor is very efficient, compact and requires very little energy to operate.

IN THE CLAIMS:

1. A catalytic selective oxidation reactor, comprising a cylindrical reactor vessel ~~(1)~~ with counter current cooling means ~~(5,6,7)~~ and at least one stage, each stage being provided with an inlet for a first feedstock ~~(2)~~, and inlet for a second feedstock ~~(8)~~, gas mixing means ~~(9)~~ and a catalytic reaction zone ~~(4)~~, wherein the reaction zone comprises a selective oxidation catalyst deposited upon a metal support.

3. A reactor according to ~~any one of the preceding claims~~ claim 1, wherein the metal support is a metal monolith.

4. A reactor according to ~~any one of the preceding claims~~ claim 1, wherein the cooling means comprises a central jacket mounted within the reactor.

6. A reactor according to claim ~~4 or 5~~, wherein the central jacket is connected via a coolant circuit to a circulating pump.

7. A reactor according to ~~any one of the preceding claims~~ claim 1, wherein the catalytic reaction zone(s) is/are annular and surround a central cooling means.

8. A reactor according to ~~any one of the preceding claims~~ claim 1, having from 2 to 10 stages.

10. A reactor according to ~~any one of the preceding claims~~ claim 1, wherein the gas mixing means comprise annular mixing vanes or discs.

11. A reactor according to ~~any one of the preceding claims~~ claim 1, connected to the output from a reformer such that CO is removed from the output gas to a level where the output from the reactor may be fed to a fuel cell.

14. A process according to claim 12 ~~or 13~~, providing hydrogen to a fuel cell.

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REACTOR AND PROCESS FOR REMOVAL OF CARBON MONOXIDE FROM HYDROGEN

The present invention concerns an improved reactor, more particularly it concerns a reactor particularly adapted for and suitable for certain selective oxidation reactions.

We have previously developed certain selective oxidation processes for the removal of residual carbon monoxide in hydrogen-containing gases intended as feedstock for fuel cells. Such processes are disclosed and claimed in for example, PCT/GB98/02873. It has become well established that hydrogen-fuelled fuel cells require very low proportions of carbon monoxide, which acts as a catalyst poison and degrades performance of the fuel cell. Desirably, the amount of CO is less than 10 ppm. If a liquid fuel is reformed to form a hydrogen-containing fuel, levels of CO are produced which may vary about 0.5-5mol%, so that it becomes necessary to attenuate such CO levels. As mentioned in the above International Application, catalytic selective oxidation reactions are highly exothermic with the result that increasing reaction temperatures lead to increasing loss of valuable hydrogen due to competing reactions.

There remains a need for a catalytic selective oxidation reactor which permits good temperature control yet is simple, effective and inexpensive to manufacture. Although hydrogen "cleaned-up" by the reactor of the present invention may be used as fuel cell fuel, there are many other uses for such hydrogen. Also, although the reactor is particularly suitable for the cleaning up of reformate (which may be from methanol, other oxygenated fuels, gasoline or other hydrocarbon fuels), it is not limited to such use. Accordingly, the present invention provides a reactor for catalytic selective oxidation, comprising a reactor vessel with counter current cooling and at least one stage, each stage provided with an inlet for a first feedstock, e.g. CO contaminated hydrogen, an inlet for a second feedstock, e.g. oxygen-containing gas, gas mixing means and a catalytic reaction zone, preferably comprising a selective oxidation catalyst deposited on a substrate having good heat conduction properties.

The invention also provides a process for the removal of quantities of CO from a hydrogen feedstock by partial oxidation, comprising at least one step of admixing said

feedstock with a quantity of oxygen, passing the resulting mixture over a selective oxidation catalyst in a selective oxidation zone and recovering a hydrogen product with reduced content of CO, whilst simultaneously cooling the selective oxidation zone, preferably to maintain the selective oxidation zone at a temperature in the range 100 to 250°C, by counter-current coolant flow.

The reactor, in its most preferred embodiment, has four stages and this is effective to reduce a reformate having a CO content of 1 to 2% to below 10ppm. In other embodiments, there may be 2 to 10 stages.

The reactor is suitably in the form of a vessel in which the reaction zones are generally annular in shape, and provided with internal and/or external liquid cooling. Cooling is preferably provided by internal cooling using water, which is suitably under pressure to prevent boiling at the temperatures generated. Other liquid coolants, or forced air cooling may be used if desired. According to specific reactor designs, heat removal may be enhanced by increasing heat exchange surface areas by corrugation or other established means. Heat removal may be altered by altering the flow of coolant and/or altering the quantity of heat removed from the coolant by means of a heat exchanger.

Each reaction zone suitably comprises a metallic catalyst support ("monolith") of generally honeycomb construction, as is in common use in automobile exhaust gas catalysis. These may have 50 to 1200 cells per square in cell density, preferably 200 to 600 cpsi, and may be of a variety of types of stainless steel. In general, the present invention does not result in the reaction stages reaching the higher temperatures generally met with in exhaust gas catalysis (e.g. 500-900°C), but the material should be capable of resisting temperature excursions. An alternative reaction zone design utilises what has become known as "static mixers" which combine a very high degree of gas agitation and mixing, resulting in non-laminar flow, with relatively low pressure drop. Such static mixers may be manufactured from a variety of metals and are commercially available. However, it is presently preferred to use a honeycomb monolith.

The reaction zone desirably carries a selective oxidation catalyst comprising a platinum group metal, especially comprising platinum and/or rhodium, carried on a high

surface area metal oxide support, such as alumina. The metallic catalyst support may be coated by generally known methods with a slurry of oxide support, dried and fired, and then impregnated with the catalytically active components, before a final firing. Such procedures are in themselves known, and do not form part of this invention.

The invention will now be described with reference to the accompanying drawing which shows a cross-section of one embodiment of the present invention.

An essentially cylindrical metal vessel, made by pressing two halves and seam-welding, has a main gas inlet, 2, for reformate gas, and an outlet, 3, for cleaned gas. The vessel has a number of annular reaction zones, 4, each consisting of an annular wound corrugated catalyst support. The vessel further has an internal water jacket, 5, between a spacer and the shell and this is supplied with cooling water by means of a water circuit comprising a circulating pump, 6, and a heat exchanger, 7. The heat exchanger is desirably used to recover heat in an integrated system e.g. in an integrated combined heat and power system. The vessel further is provided with four air inlets, 8, each providing pre-determined (for a steady-state operation) levels of air (that is reaction oxygen; air is generally suitable). Although not preferred, controlled amounts of oxygen, according to one or more feedback or feed forward controls detecting gas flow levels and CO levels may be used. Downstream of each air inlet is a gas mixing zone, 9a, 9b, 9c and 9d. Zone 9a is suitably a static mixer, but zones 9b, 9c and 9d are preferably alternating annular mixing vanes, 10, and discs, 11, having gas flow holes, as illustrated in Figs 2 and 3. It should be noted that for simplicity only four vanes and four holes are shown, but more, e.g. six or eight, may be used. Such vanes and discs may be manufactured by pressing to form a sleeve which may be fitted and brazed or welded onto the jacket 5 with the intention that each gas mixing zone also serves to remove hear from the reaction gases leaving each reaction zone. Although the schematic drawing of the reactor shows each reaction zone and gas mixing zone of the same size, these may be designed to differ according to the amount of heat to be removed. Additionally, each reaction zone may incorporate different and/or different loadings, of catalyst. Each metal catalyst support monolith suitably has 300-400 cells per sq.in. and is coated with a washcoat comprising ~36wt% alumina solids of D50 of approx 5μm and D90 of approx 16μm, to deposit approx $3g/m^3$. After drying and firing, the coated monolith was impregnated with

a platinum salt solution to result in 5% by wt of Pt deposited on the alumina. The resulting coated monolith was again dried and fired before being assembled into the reactor.

Initial studies indicate that a reformate containing 1-2% CO in admixture with H_2, CO_2, N_2, water vapour with a small proportion of unreacted reformer feedstock (e.g. CH_3OH, CH_4, gasoline), at 180°C may be converted to a feed gas suitable for a fuel cell, containing below 10ppm CO, and at 140°C.

The reactor of the invention is compact and exhibits relatively low pressure drop. Manufacturing costs are relatively low. The reactor is believed to be suitable for mobile or automobile uses as well as stationery uses. It is presently envisaged that the reactor shell will be lagged to control heat loss and prevent injury. The reactor can be manufactured simply and inexpensively, using pressed and welded parts. It is desirably operated to achieve low pressure drop, and the particular embodiment illustrated is designed for a gas hourly space velocity of approx. 30,000h^{-1} gas throughput.

The reactor as described may be altered in a variety of ways without departing from the central inventive concept.

The invention is now illustrated below in a working example of a two-stage reactor, fed with a synthetic reformate gas mixture containing N_2, H_2, CO, CO_2 and H_2O. Each of the two stages consists of a mixing zone with three off-set spiral vanes, followed by air injection accomplished through a number of apertures in an annular ring, producing a swirling, mixing gas. The thus mixed hydrogen and air mixture passes through an annular metal honeycomb support mounted on a central cooling jacket. The cooling jacket is fed counter-current to the flow of hydrogen with a heat transfer fluid, conveniently water under pressure, and the metal honeycomb support is mounted in good heat exchange contact with the cooling jacket. The metal honeycomb carries a selective oxidation catalyst. Gas may be extracted for analysis between the two stages and/or after the second stage. By setting the composition of the feedstock gas mixture according to that analysed after the first or second stage, the test two-stage reactor can model a three or four stage reactor.

The detailed operating conditions and other experimental detail for one typical run of the above reactor now follow. It was remarkable that the pressure drop across the whole of the est reactor was about the measurement limit of the gauges being used, and was of the order of 1.5-3mbar (150-300Pa), at the two throughputs tested. This indicates that the reactor is very efficient, compact and requires very little energy to operate.

1. Each catalyst support was an annular of internal diameter 3.85in (9.78cm), external diameter 5.85in (14,86cm) and 3.0in(7.62cm) length, made from "Fecralloy" steel and having 500 cells per sq in. Each support had been washcoated with alumina washcoat as used in exhaust gas catalytic convertors, followed by loading with 5% by wt of the alumina of Pt. Conventional manufacturing techniques were used.

2. The synthetic reformate had the compositions: 48%N_2(dry), 40%H_2 (dry), 10%CO_2(dry), 0.5-1.5%CO (dry) and 13% H_2O (of total flow). Two levels of CO concentration were chosen, a higher level to represent the output from autothermal reforming, and a lower level to represent the output from steam reformer or an optimised reformer, and two levels of throughput stated to be Nominal 3kWe (1kilowatt equivalent = 1,000 l/hr of H_2) and Nominal 6kWe, equal to 7,5000 l/hr of dry reformate and 15,000 l/hr of dry reformate respectively (8,620 l/hr and 17,240 l/hr of wet reformate).

It is plain from these results with a two-stage reformer that the compact and efficient reformer of the invention will be effective to remove CO from Hydrogen to any desired level, by conventional optimisation and especially by matching the number of stages to the requirements of the fuel cell system or other usage of the purified hydrogen.

TWO-STAGE ANNULAR SELOX REACTOR TEST RESULTS

FIRST STAGE							SECOND STAGE				
Ex No.	Power kWe	CO in %, dry	Air in, slm	Temp in C	Temp out, C	CO out %, dry	Air in, slm	Temp in, C	Temp out, C	CO out ppm dry	Sel**
1	3	1.5	4	125	212	0.60	2	167	212	1610	0.696
2	3	1.5	4	127	214	0.60	3	167	217	740	0.640
3	3	1.5	4	125	210	0.60	4	169	227	950	0.556
4	3	1.5	3	125	149	0.81	3	129	175	1840	0.684
5	3	1.5	5	128	215	0.34	3	161	220	290	0.582
6	3	0.5	2	143	176	0.122	0.5	138	145	232	0.579
7	3	0.5	2	141	167	0.122	0.8	127	136	29	0.540
8	3	0.5	2	140	161	0.122	1.0	121	130	28	0.505
9	3	0.5	2	141	172	0.122	1.2	131	146	55	0.472
10	3	0.5	1.5	142	166	?	1.0	134	157	373	0.562
11	3	0.5	2.5	142	179	?	1.0	138	150	50	0.433
12	6	0.5	4	138	167	0.130	1	148	176	722	0.520
13	6	0.5	4	138	162	0.130	2	140	185	51	0.503
14	6	0.5	4	138	166	0.130	3	146	198	105	0.428
15	6	0.5	6	138	205	62ppm	0	176	174	62	0.502

** Sel: (Selectivity) = CO consumed/2 x O_2 consumed

CLAIMS

1. A catalytic selective oxidation reactor, comprising a cylindrical reactor vessel (1) with counter current cooling means (5,6,7) and at least one stage, each stage being provided with an inlet for a first feedstock (2), and inlet for a second feedstock (8), gas mixing means (9) and a catalytic reaction zone (4), wherein the reaction zone comprises a selective oxidation catalyst deposited upon a metal support.

2. A reactor according to claim1, wherein the metal support is mounted in good heat exchange contact with the cooling means.

3. A reactor according to any one of the preceding claims, wherein the metal support is a metal monolith.

4. A reactor according to any one of the preceding claims, wherein the cooling means comprises a central jacket mounted within the reactor.

5. A reactor according to claim 4, wherein the central jacket is connected via a coolant circuit to an external heat exchanger.

6. A reactor according to claim 4 or 5, wherein the central jacket is connected via a coolant circuit to a circulating pump.

7. A reactor according to any one of the preceding claims, wherein the catalytic reaction zone(s) is/are annular and surround a central cooling means.

8. A reactor according to any one of the preceding claims, having from 2 to 10 stages.

9. A reactor according to claim 8, having four stages.

10. A reactor according to any one of the preceding claims, wherein the gas mixing means comprise annular mixing vanes or discs.

11. A reactor according to any one of the preceding claims, connected to the output from a reformer such that CO is removed from the output gas to a level where the output from the reactor may be fed to a fuel cell.

12. A process for the removal of quantities of CO from a hydrogen feedstock by partial oxidation, comprising at least one step of admixing said feedstock with a quantity of oxygen, passing the resulting mixture over a selective oxidation zone comprising a selective oxidation catalyst on an annular metal support, and recovering a hydrogen product with reduced content of CO, whilst simultaneously cooling the selective oxidation zone by counter-current coolant flow.

13. A process according to claim 12, wherein the selective oxidation zone is maintained at a temperature in the range 100 to 250°C.

14. A process according to claim 12 or 13, providing hydrogen to a fuel cell.

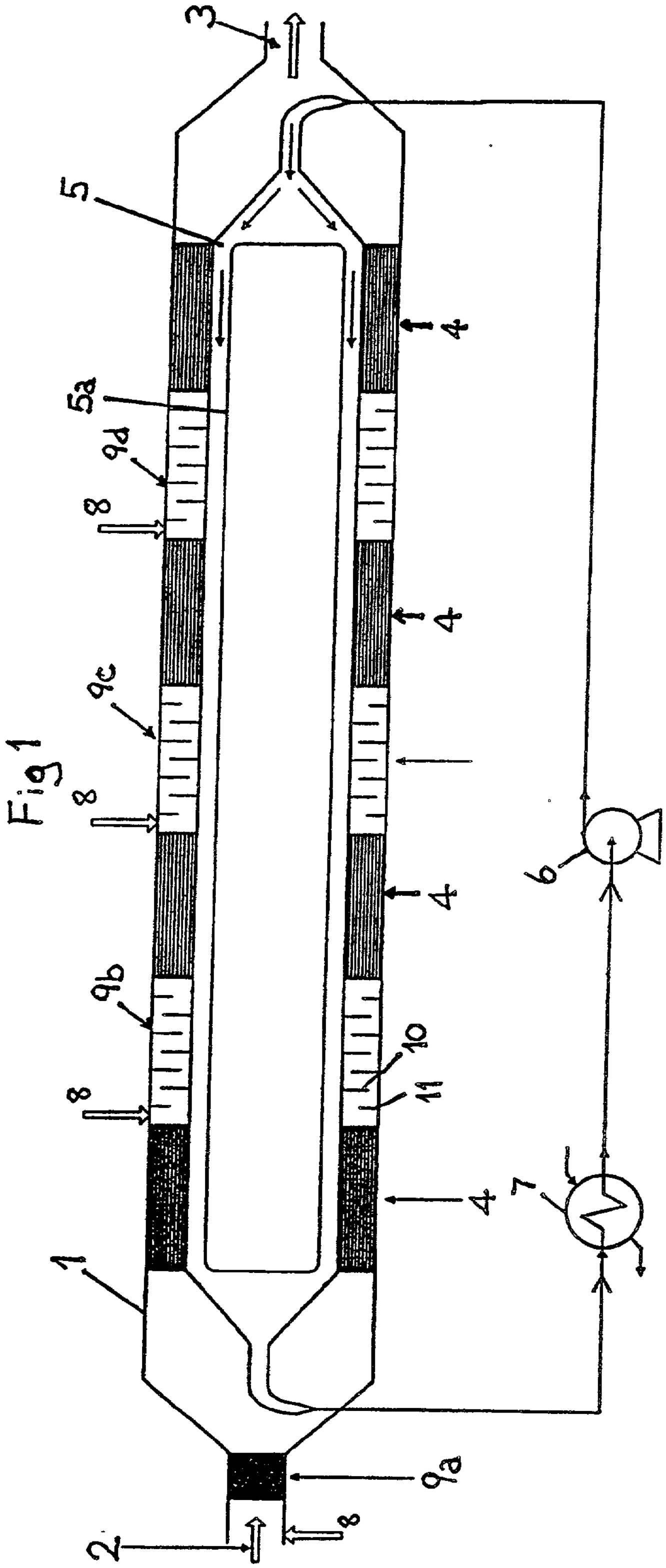

Fig 1

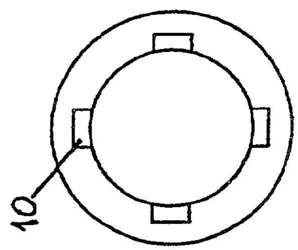

Fig 3

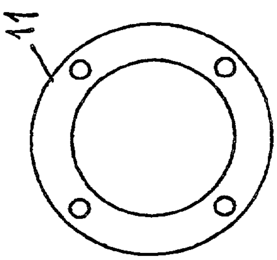

Fig 2

Declaration and Power of Attorney For Patent Application
English Language Declaration

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name,

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled
REACTOR AND PROCESS FOR REMOVAL OF CARBON MONOXIDE FROM HYDROGEN,
the specification of which is attached hereto unless the following box is checked:

☒ was filed on June 1, 2001 as
United States Application Number or PCT International Application Number 09/857,116
and was amended on June 1, 2001.

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR § 1.56.

I hereby claim foreign priority benefits under 35 U.S.C. §119(a)-(d) or § 365(b) of any foreign application(s) for patent or inventor's certificate, or § 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed:

Prior Foreign Application(s)			Priority Not Claimed
9826222.3	Great Britain	01 December 1998	
(Number)	(Country)	(Day/Month/Year Filed)	☐
_____	_____	_____	
(Number)	(Country)	(Day/Month/Year Filed)	☐

I hereby claim the benefit under 35 U.S.C. § 119(e) of any United States provisional application(s) listed below.

_____	_____
(Application Number)	(Filing Date)
_____	_____
(Application Number)	(Filing Date)

I hereby claim the benefit under 35 U.S.C. § 120 of any United States application(s), or 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of 35 U.S.C. § 112, I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR § 1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application:

(Application Number) (Filing Date) (Status - patented, pending, abandoned)

(Application Number) (Filing Date) (Status - patented, pending, abandoned)

POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith:

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Paul F. Prestia	Reg. No. 23,031	Lawrence E. Ashery	Reg. No. 34,515	Jack J. Jankovitz	Reg. No. 42,690
Allan Ratner	Reg. No. 19,717	Christopher R. Lewis	Reg. No. 36,201	Jonathan H. Spadt	Reg. No. 45,122
Andrew L. Ney	Reg. No. 20,300	Robert L. Andersen	Reg. No. 25,771	Christopher I. Halliday	Reg. No. 42,621
Kenneth N. Nigon	Reg. No. 31,549	Joshua L. Cohen	Reg. No. 38,040	Scott A. Mckeown	Reg. No. 42,866
Kevin R. Casey	Reg. No. 32,117	Daniel N. Calder	Reg. No. 27,424		
Benjamin E. Leace	Reg. No. 33,412	Louis W. Beardell, Jr.	Reg. No. 40,506		
James C. Simmons	Reg. No. 24,842	Jacques L. Etkowicz	Reg. No. 41,738		

Address all correspondence to: Christopher R. Lewis
Ratner & Prestia, Suite 301, One Westlakes, Berwyn, P.O. Box 980, Valley Forge, PA 19482-0980
Address all telephone calls to: Christopher R. Lewis at (610) 407-0700.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

1-00

Full name of sole or first inventor (given name, family name) Peter Geoffrey Gray

Inventor's signature P Gray Date 24 July 01

Residence Reading, United Kingdom GBX

Citizenship Australian

Post Office Address The Linney, Blounts Court Road, Sonning Common
Reading RG4 9RS, United Kingdom

2-00

Full name of second joint inventor, if any (given name, family name) John Frederick Pignon

Second Inventor's signature J. F. Pignon. Date 30 - 7 - 01

Residence Oxon, United Kingdom GBX

Citizenship British

Post Office Address 14 Castle Close, Benson
Oxon OX10 6SN, United Kingdom

☐ Additional inventors are being named on separately numbered sheets attached hereto.